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Cryogenic-based CO_2 capture technologies: State-of-the-art developments and current challenges

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ABSTRACT

CO_2 capture, utilization and storage has been recognized as a primary option to mitigate the issue of climate change caused by the utilization of fossil fuels. Several CO_2 capture strategies have been developed, such as absorption, adsorption, membrane, chemical looping, hydrating and biofixation. Among different technologies, particular attention has been given to cryogenic CO_2 capture by phase change. The aim of this study is to improve interest in cryogenic technologies for CO_2 capture by providing an overview of the actual status of CCS. To reach this goal, the major strategies and technologies for CO_2 capture from fossil fuel combustion have been reviewed. Simultaneously, the characteristics of cryogenic technologies for CO_2 capture are summarized. The existing challenges that need to be overcome in cryogenic technology include cold energy sources, capture costs and impurities, etc. Finally, opportunities for the future development of cryogenic-based technologies are discussed. The results of this investigation indicated that cryogenic CO_2 capture processes can be easily retrofitted to the existing industrial emission facilities and avoid the challenges associated with chemical solvents or physical sorbents.

1. Introduction

Recently, energy and environmental issues have attracted worldwide concern. Although more energy is being developed from non-fossil energy sources, the main energy supply will still come from fossil fuels [1]. Consequently, there is an urgent demand for the use of reasonable and sustainable fossil fuels with decreased environmental consequences [2]. As one of the most important greenhouse gases, CO_2 generated from fossil fuel combustion plays an important role in climate change. Among different emission sources, coal-fired power plants are one of the most dominant CO_2 emission sources throughout the world and discharge approximately 2 billion tons of CO_2 per year, as shown in Fig. 1 [3,4]. In the last decades, the global CO_2 emission has rapidly increased, especially in developing countries (i.e., China and India), as shown in Fig. 2 [4]. According to a report by the Intergovernmental Panel on Climate Change, by the year 2100, the atmosphere may contain up to 570 ppm of CO_2, causing a rise in the mean global temperature of approximately 1.9 °C and a 3.8 m increase in the average sea level [5].

CO_2 capture, utilization and storage (CCUS) is a promising strategy to mitigate CO_2 emission by capturing it from large point sources (e.g., coal-fired power plants, iron-steel and cement industry, etc.), storing it in porous rocks (usually lower than 700 m underground), or reusing it for oil recovery, fertilizers and chemical synthesis, etc. [7]. By retrofitting CO_2 capture and storage units into a conventional power plant, CO_2 emissions could be reduced by approximately 80–90% [5]. Different CO_2 capture and separation technologies have been widely researched by scientists, and the essential issue is the development of novel materials or structures that will allow the minimum energy penalty and an optimal capture efficiency [8–12].

Although there are several choices for CO_2 capture from large emission sources (i.e., absorption, adsorption, membrane and cryogenic, etc.), the selection of a suitable capture approach is highly dependent on specific discharge conditions [13]. The primary criterion is the state of the flue gas, including its composition, temperature, flow rate, and CO_2 concentration (also namely, the CO_2 partial pressure). In

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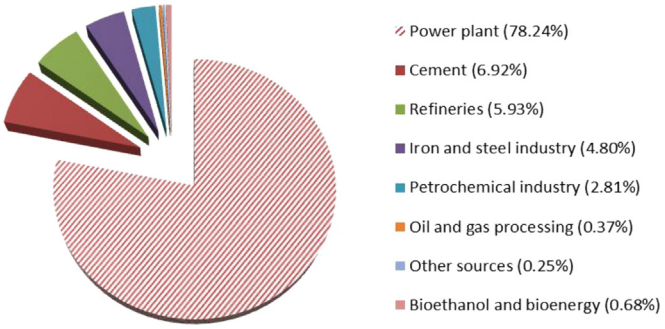

Fig. 1. Breakdown of the dominant CO_2 emission sources [6].

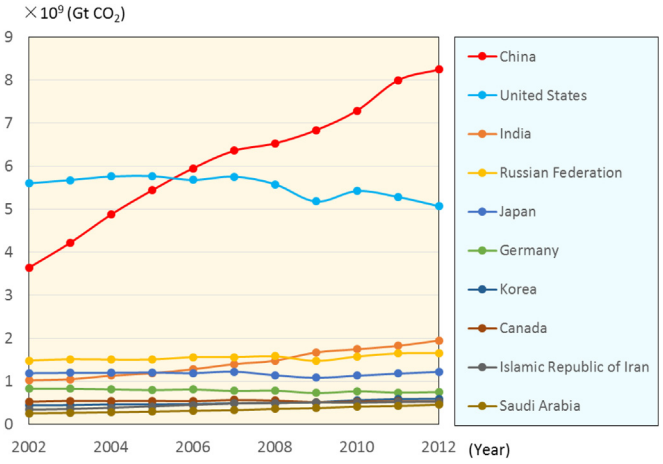

Fig. 2. CO_2 annual emission from fuel combustion of the top ten countries in the world [4].

addition, the specifications for targeted production (e.g., purity of CO_2 and transport pressure) or discharged standards (e.g., H_2S, SOx and NOx, etc.) also have a significant influence on the selection of a capture technology [14]. For example, to facilitate transportation and storage, the captured CO_2 should be compressed to over 110 bar [15,16].

One traditional method is the application of cryogenic technologies for CO_2 separation, which has increased in recent years due to its advantages including avoiding the use of chemical solvents and no secondary pollution [17–19]. To use cryogenic technologies in an efficient way, a review of the advantages and challenges in the existing processes is necessary. In this work, the potential for and applications of cryogenic separation technologies in CO_2 capture were explored, and general guidelines on some design parameters used to choose a suitable technology for CO_2 separation are discussed. The relationship between the low temperature characteristics and the main variables affecting the performance of each technique were investigated. The paper is organized as follows: Section 2 summarizes existing CO_2 capture strategies and technologies. Section 3 introduces existing low-temperature CO_2 capture technologies. Sections 4 and 5 discuss the advantages, challenges and potential solutions for low-temperature processes. Section 6 predicts the further direction of low-temperature CO_2 capture technologies. Section 7 summarizes the main conclusion of this work.

2. Existing CO_2 capture strategies and technologies

2.1. Existing CO_2 capture strategies

The main strategies for CO_2 capture from fossil fuel combustion processes consist of pre-combustion capture, oxy-fuel combustion capture and post-combustion capture (as shown in Fig. 3) [20]. The selection of a CO_2 capture strategy is mainly based on its advantages and disadvantages, and the CO_2 emission conditions (e.g., the concentration of CO_2 in the flue gas, the pressure of the gas stream), as shown in Tables 1 and 2 [20].

2.1.1. Pre-combustion CO_2 capture

In the pre-combustion capture process, fuel reacts with O_2 and H_2O. During the reaction, the carbon in the fuel is converted to CO_2 and CO, and simultaneously, H_2 is produced. After the water-gas shift (WGS) reaction, CO is converted into CO_2, and the main components in the mixed gas (namely, syngas) are approximately 60–80% H_2 and 20–40% CO_2 [21]. This capture strategy is mainly used for coal gasification in an integrated gasification combined cycle (IGCC). The significant advantage of pre-combustion is that the produced H_2 is an ideal green energy source because its combustion emits no waste gas and only generates water [22]. Meanwhile, H_2 can be used in many areas, such as in fuel batteries, new energy vehicles, the aerospace industry, chemical industry, etc. [23].

2.1.2. Oxy-fuel combustion CO_2 capture

In the oxy-fuel combustion CO_2 capture process, fuel combusts in pure O_2, causing a high CO_2 concentration (over 80%) in the flue gas, and thus, the purification of CO_2 in this process is easier than that in post-combustion strategies [24,25]. Moreover, the NOx content in the flue gas is also lower with oxy-fuel combustion. Due to the combustion taking place in pure O_2, the high temperature flue gas is often recycled to the combustor to recover waste heat. Currently, oxy-fuel combustion is usually associated with pulverized coal combustion, and fluidized bed (FB) combustors could be a promising alternative technology. The main advantage of a fluidized bed combustor over pulverized coal oxy-fuel combustion is its ability to reduce the flue gas flow for a given coal input while maintaining a high furnace temperature [25]. A crucial challenge is that the efficiency of coal-fired power plants would obviously be reduced by 10–12% if oxy-fuel CCS utilities were retrofitted to it, due to the air separation unit (ASU) and compression and purification unit (CPU) [26]. To capture CO_2 in an efficient way by oxy-fuel combustion, the energy penalty caused by the need for an ASU and CPU must be further minimized via optimization of the process and heat integration.

2.1.3. Post-combustion CO_2 capture

In the post-combustion CO_2 capture process, fuel combusts in the air. In the conventional combustion process, the concentration of CO_2 in the flue gas is low (typically 4–14%) of the atmospheric pressure [20]. For this reason, there is a rigorous demand for energy and equipment to enable CO_2 separation and achieve the required CO_2 concentration (above 95.5%) for transport and storage [27]. Compared with pre-combustion and oxy-fuel combustion, post-combustion CO_2 capture units can be directly added to existing coal-fired power plants with little retrofitting, and this method has been demonstrated at some pilot-scale sites. As one of the most mature post-combustion CO_2 separation technologies, the energy requirements for solvent regeneration in the monoethanolamine (MEA) absorption process vary from 3.0 to 4.5 MJ/kg CO_2, which contributes to approximately 80% of the total energy consumption [28,29]. In contrast, a target of 2 MJ/kg CO_2 (including both the capture and compression steps) is often mentioned in the European Union recommendations [30,31].

2.2. Existing CO_2 capture technologies

Currently, there are several CO_2 separation technologies in use such as absorption, adsorption, membrane and cryogenic, etc. Each of these technologies is based on different separation principles. For this reason, the selection of appropriate technologies for various industrial emission sources is important in order to achieve high efficiencies, and the efficiency is dependent on different parameters (e.g., stream conditions, flue gas composition, economics and target products) [40].

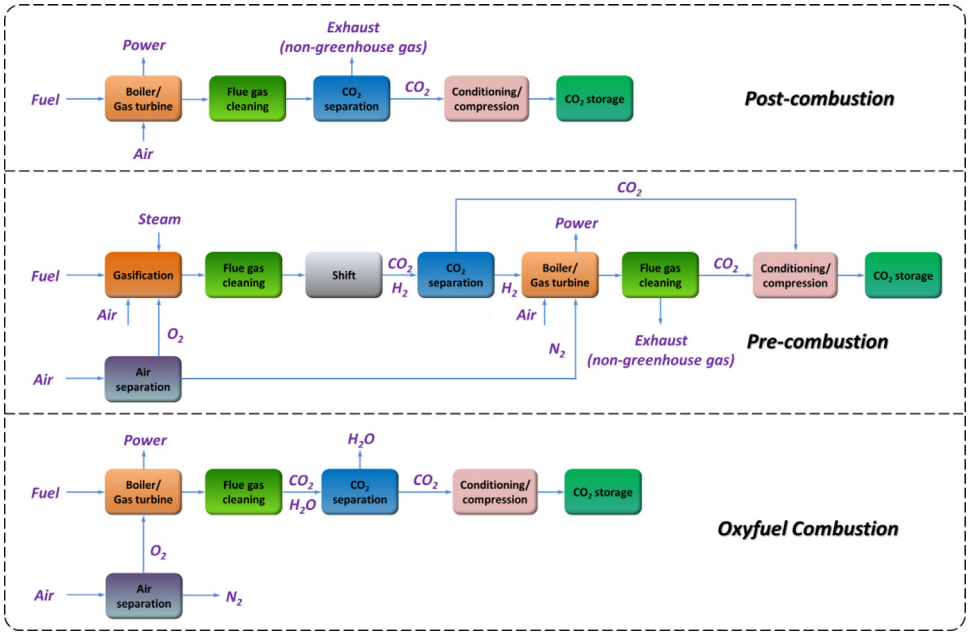

Fig. 3. The dominant CO_2 mitigation strategies for fossil fuel combustion.

Table 1
The advantages and disadvantages of the existing CO_2 capture strategies.

CO_2 capture strategies	Advantages	Disadvantages	Ref.
Pre-combustion	• High CO_2 concentration (~45 vol%) and pressure • Commercial applied in some industrial sectors	• Severe operating conditions (15–20 bar and 190–210 °C) • Energy penalty due to sorbent regeneration	[32,33]
Oxy-fuel combustion	• Lower capital cost • High CO_2 concentration (80–98%) • Low investment of boiler and other equipments	• High efficiency drop and energy penalty due to ASU	[34,35]
Post-combustion	• A straightforward approach to be retrofitted • More mature than other strategies	• Dilute CO_2 concentration (5–15 vol%) at near atmospheric pressure • Energy penalty due to solvent/sorbent regeneration	[27,34]

Table 2
The flue gas composition of different CO_2 capture strategies.

Composition of flue gas	Pre-combustion [36,37]	Oxy-fuel combustion [13,38,39]	Post-combustion [36,37]
CO_2	37.7%	85.0%	10–15%
N_2	3.9%	5.8%	70–75%
H_2O	0.14%	100 ppm	5–10%
H_2	55.5%	–	–
O_2	–	4.7%	3–4%
CO	1.7%	50 ppm	20 ppm
NOx	–	100 ppm	< 800 ppm
SOx	–	50 ppm	< 500 ppm
H_2S	0.4%	–	–

2.2.1. Absorption

Monoethanolamine (MEA) absorption is the most common technology used for post-combustion CO_2 capture, including two sections of absorption and stripping/desorption, as shown in Fig. 4. It is capable of achieving high-level CO_2 capture (more than 90%) from flue gas via fast kinetics and strong chemical reactions. However, there are several remaining limitations that need to be solved including solvent degradation, corrosion, and solvent regeneration efficiency [129]. The amines are corrosive and susceptible to degradation by trace constituents (particularly SOx), which seriously restricts the application of this technology. According to work by Rao and Rubin [41], solvent degradation contributes to approximately 10% of the total CO_2-capture costs. There are two main types of degradation: 1) thermal degradation, which occurs in conditions with a high temperature and high CO_2 partial pressure. 2) Oxidative degradation, which is mainly due to the presence of a large amount of O_2 in the flue gas. Some impurity gases (e.g., SOx and NOx) can also cause solvent degradation. The volatile components could be emitted into the atmosphere and are environmentally hazardous. Moreover, the regeneration of chemical solvents requires a significant amount of energy (approximately 4–6 MJ/kg CO_2) [3]. The Department of Energy/National Energy Technology Laboratory (DOE/NETL) [42] reported that an MEA-based process for CO_2 capture will increase the cost of electricity for a new power plant by approximately 80–85% and reduce the plant efficiency by approximately 30%.

2.2.2. Adsorption

Fig. 5 illustrates a typical configuration for physical CO_2 adsorption. As shown in the figure, a pretreatment stage is necessary prior to CO_2 adsorption. Each of the adsorption chambers is packed with a solid adsorbent (i.e., activated carbon, zeolites, or metal organic frameworks, etc.) [43,44]. Usually, there are two or three adsorption chambers throughout the whole process, such that, during operation, one chamber is receiving the feed for adsorption, a second chamber is desorbing the captured CO_2, and the third chamber is on stand-by to receive the feed [130]. Thus, the system can operate continuously.

Until now, most CO_2 adsorption technologies are assumed to be dry processes. Other limitations that make this process less effective are [45–47]: 1) low CO_2 selectivity and capacity in available adsorbents. 2) lower removal efficiencies compared to other technologies such as absorption and cryogenics. 3) regeneration and reusability of adsorbents.

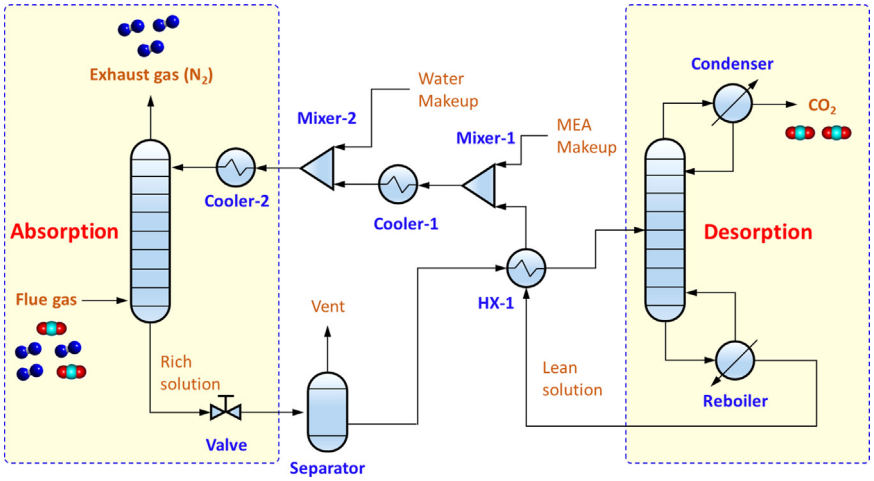

Fig. 4. Typical chemical absorption system for CO_2 recovery from flue gas.

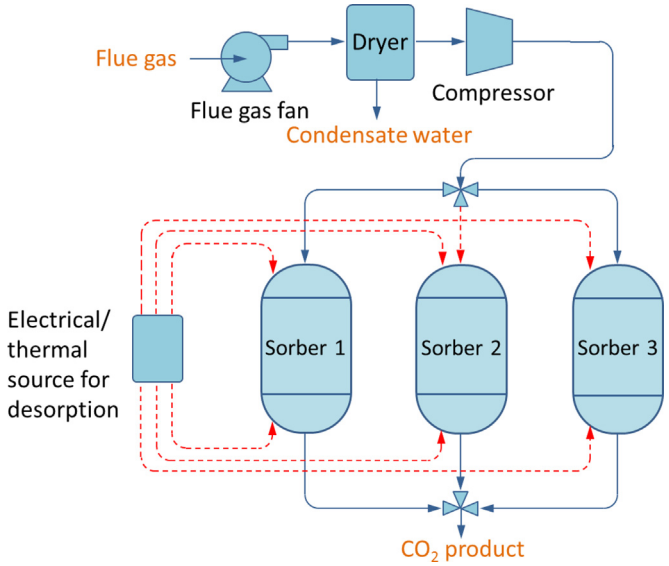

Fig. 5. Process flow diagram for the CO_2 capture from flue gas by adsorption process.

2.2.3. Membranes

Membranes are a relatively novel separation technology and have been explored for CO_2 capture in recent years. As a promising alternative, membrane-based technology has become a competitive CO_2 capture process due its simplicity, energy efficiency and eco-friendliness [48–50]. Fig. 6 depicts a two-stage membrane separation strategy using part of the permeate as a sweep, where 70% of the permeate from the first-stage membrane is fed to the second-stage membrane as a feed, and the remaining 30% of the permeate is re-circulated as a sweep to the first-stage membrane [131], whereas 5% of the permeate from the second-stage membrane is re-circulated as a sweep to the second-stage membrane. This sweep distribution corresponds to approximately 5% of the feed flow rate in each respective membrane stage [131].

Nevertheless, there are also some disadvantages that limit the ultimate use of membranes [51–53]. First, membranes have strict temperature requirements. Their structures will be destroyed rapidly once the temperature of the flue gases exceeds 100 °C [53]. Meanwhile, membranes are sensitive to corrosive gases (e.g., SOx, NOx and H_2S); thus, pretreatment prior to the membrane separation process is necessary. It is difficult to maintain membrane performance over long-term operation. Most membranes do not have resilience in practical industry conditions and quickly fail, which is one of the biggest challenges for their potential application in industrial practice [54]. Additionally, when the concentration of CO_2 in the feed stream is diluted (below 20%), multiple stages and/or recycling of one of the streams is also necessary.

2.2.4. Chemical looping cycle

Chemical-looping combustion (CLC), proposed by Richter and Knoche [128], divides combustion into intermediate oxidation and reduction reactions that are performed separately with a solid oxygen carrier circulating between the separated sections. Small particles of metal oxide, such as Fe_2O_3, NiO, CuO or Mn_2O_3 [55,56], are suitable oxygen carriers. A basic CLC system with two reactors, one each for air and fuel, is shown in Fig. 7. The oxygen carrier circulates between the reactors. In the air reactor, the carrier is oxidized by oxygen. In the fuel reactor, the metal oxide is reduced by the fuel, which is oxidized to CO_2 and H_2O. The main advantages of CLC can be summarized as [20,132]:

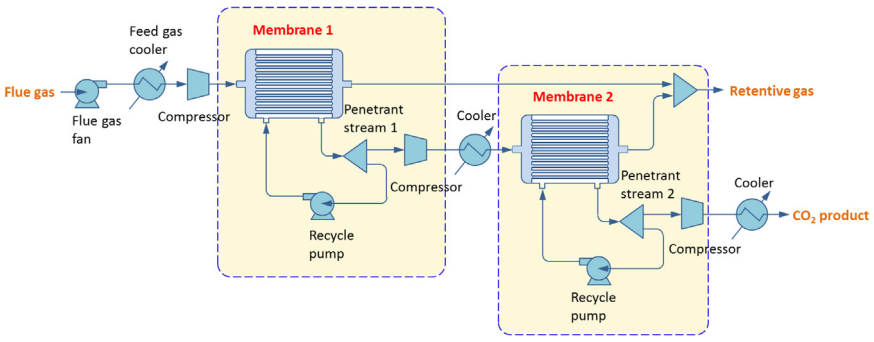

Fig. 6. Process flow diagram for the CO_2 capture from flue gas by membranes (recycled permeate).

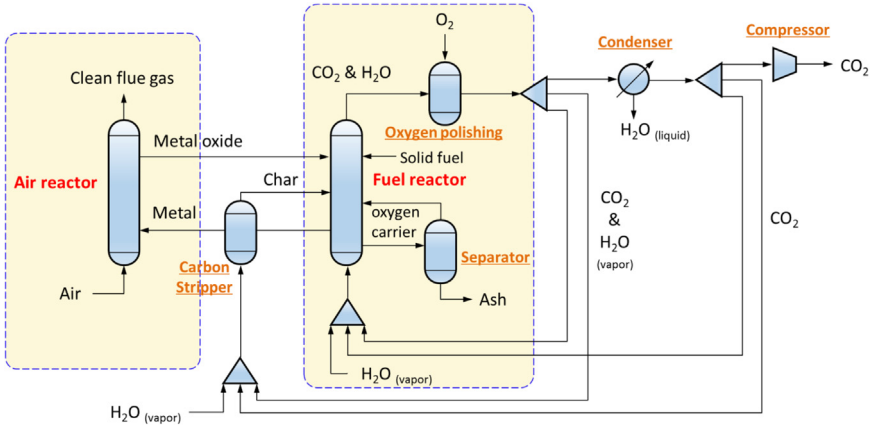

Fig. 7. Process flow diagram for the CO_2 capture from flue gas by chemical-looping combustion process.

1) the exhaust gas from the air reactor is mainly N_2, and thus, is harmless. 2) The exhaust gas stream from the fuel reactor is composed of CO_2 and H_2O; therefore, the CO_2 can be easily separated by a condenser, which avoids the energy penalty of conventional absorption processes and reduces the capital cost.

At present, most CLC processes have only been tested on the laboratory-scale, and there are few large-scale demonstrations of this technology. Meanwhile, several significant problems still remain in the existing processes (such as insufficient stability of the oxygen carrier and slow redox kinetics) [57]. In addition, desulfurization of the fuel is also necessary in order to avoid sulfidation of the carrier.

2.2.5. Microalgae

Currently, CO_2 bio-fixation via microalgae has gained a huge momentum due to its high photosynthetic rate, which allows the bio-fixation of CO_2 more efficiently than in terrestrial plants [58,59]. A summarized microalgae CO_2 fixation and biodiesel production process is introduced in Fig. 8. The capture process using microalgae has the following advantages [60,61,133]: 1) It is an environmentally sustainable method; 2) It directly uses solar energy; 3) it co-produces high added-value ingredients from biomass, such as human food and animal feeds, mainly for aquaculture, cosmetics, medical drugs, fertilizers, biomolecules for specific applications and biofuels.

Due to the lack of understanding regarding the microalgae CO_2 fixation process, several issues are normally ignored (such as the requirement for an inorganic nutrient source and the intensive energy used in cultivating, harvesting and drying microalgae biomass) [62–64]. The consequences of these limitations may lead to a negative CO_2 and energy balance for the life-cycle of microalgae biodiesel production. Apart from that, the low solubility of CO_2 in water is another problem that needs further attention.

3. Low temperature CO_2 capture technologies

Cryogenic technologies separate CO_2 gas from flue gases using their different condensation and desublimation properties. This method can obtain higher CO_2 recovery (99.99%) and purity (99.99%) than other separation technologies [3]. Although there may be a risk of blockages from other components (e.g., water) and increasing capture costs, more and more attention has been placed on cryogenic-based approaches [18]. Until now, several types of cryogenic CO_2 capture processes have been reported. Different targeted solutions have been proposed, such as advanced cryogenic separation systems that avoid blockages from condensed water and enhanced thermal efficiency that decreases the exergy loss and energy penalties.

3.1. Cryogenic packed bed

Tuinier et al. [65] proposed cryogenic CO_2 capture using dynamically operated packed beds, as shown in Fig. 9. The packed material is a steel monolith structure, and the cold energy is provided by liquefied natural gas [17]. The advantage of this method is that both H_2O and CO_2 can be simultaneously separated from the flue gas based on the differences in their dew and sublimation points. Issues of clogging and pressure drops can be avoided in the cryogenic packed bed. Furthermore, a chemical absorbent and elevated pressure are not required because the amount of frosted CO_2 (a volume fraction of 0.06) is determined by the cold energy stored in the packing material, and is far less than the gas void fraction of a packed bed (volume fraction of 0.4) [66].

In addition to CO_2 capture from flue gas, the cryogenic packed beds can also be utilized to upgrade biogas [67]. Similarly, CO_2 and H_2S can be deposited on the surface of the packing bed at different positions on the bed. Meanwhile, high purity CH_4 (99.1%) gathers at the exit without undergoing a phase change. Compared to a conventional vacuum pressure swing adsorption (VPSA) process, the CH_4 recovery and productivity of the cryogenic method can be improved to 94.3% and 350.2 kg CH_4 h^{-1} $m_{packing}{}^{-3}$ (79.7% and 43.1 kg CH_4 h^{-1} $m_{packing}{}^{-3}$ for the VPSA process), respectively [67]. The other advantage of the cryogenic packed bed is that the installation investment is lower than that of the VPSA process due to its smaller bed size. Moreover, the energy consumption is 22% lower (2.9 MJ/kg CH_4) than that of the VPSA process (3.7 MJ/kg CH_4) [67]. If the CH_4 product needs to be liquefied and injected into a pipe network, the cryogenic packed bed also shows a unique advantage, since the CH_4 stream leaving the packed beds is already at a cryogenic temperature, and thus, part of liquefaction costs can be avoided.

Although the cryogenic packed-bed CO_2 capture process has more potential than MEA absorption and VPSA processes, several challenges still need to be overcome before its commercial application. Thermal insulation of the cryogenic packed beds should be improved to avoid sensible and latent heat loss. Generally, the H_2S content in raw biogas is trace (ranging from 0.0001 to 1 vol%) [68]. To achieve a high H_2S removal efficiency, the temperature of the packed bed should be kept low (around −150 °C), which may lead to increased operating costs [68]. The cold energy required in this cryogenic process is competitive when liquefied natural gas (LNG) is available as a cold source. If not, the use of a refrigerator is inevitable, the energy consumption may dramatically increase, and the energy-saving advantages of the cryogenic process would be weakened [68].

3.2. External cooling loop cryogenic carbon capture (CCCECL)

At present, the main challenge of most CO_2 capture processes is

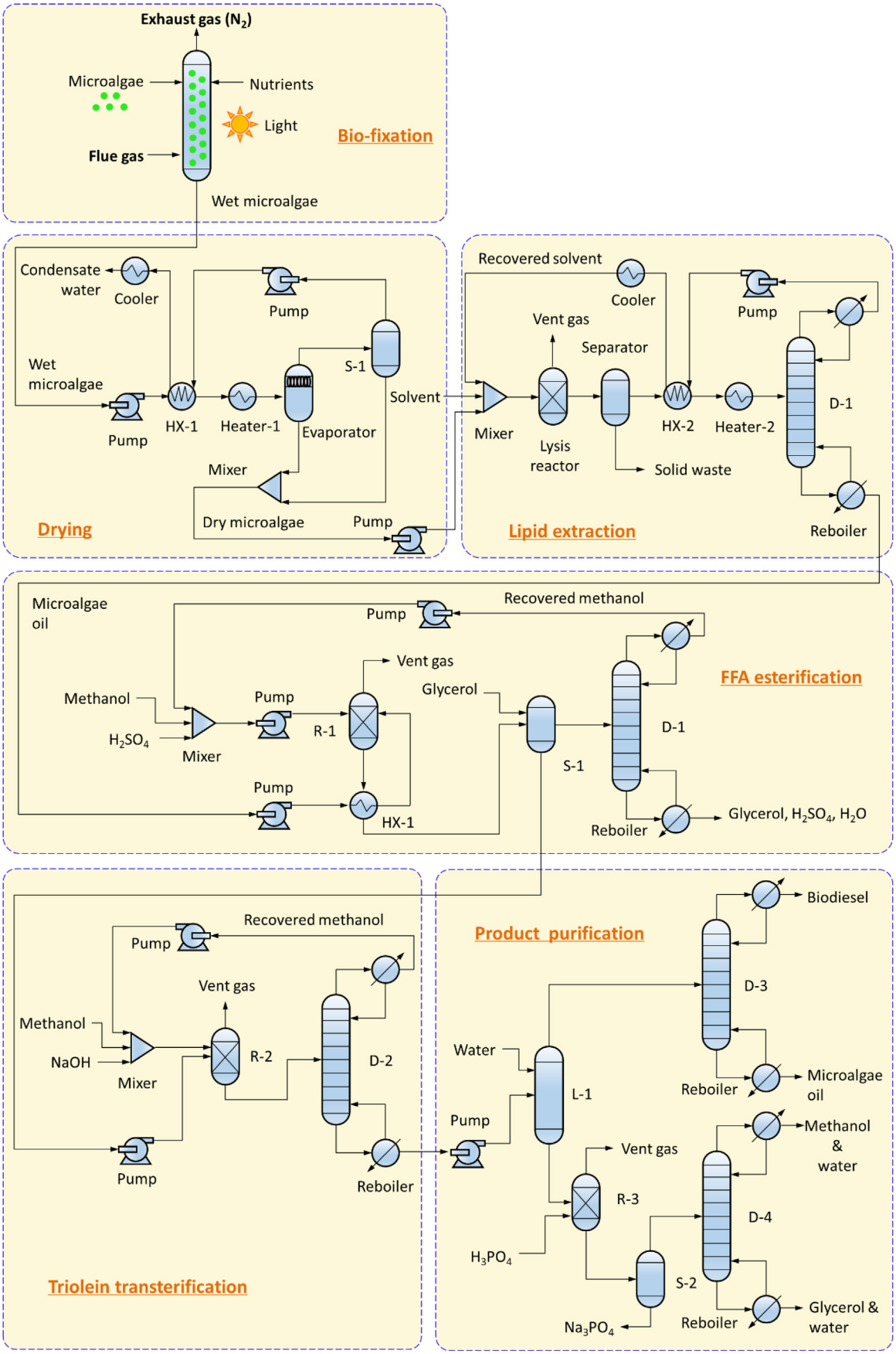

Fig. 8. Process flow diagram for the CO_2 capture from flue gas by microalgae fixation process.

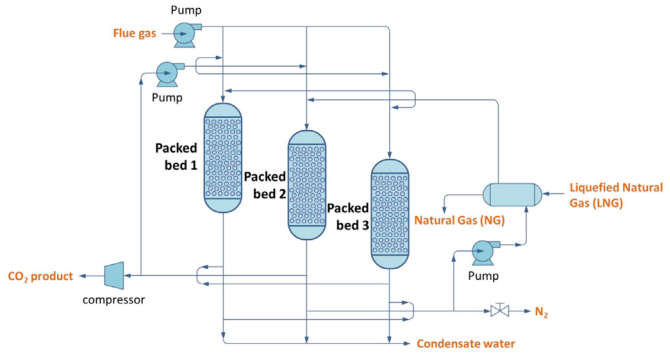

Fig. 9. The schematic diagram of low temperature CO_2 capture process by packed bed [65].

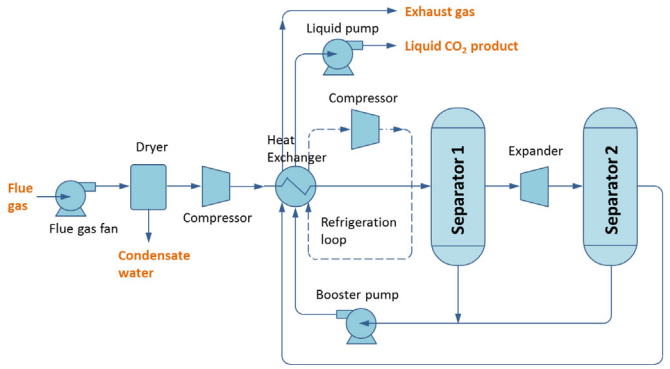

Fig. 10. Process flow diagram for the CO_2 capture from flue gas by low temperature process [69].

their high energy load. For cryogenic capture technologies, an effective approach to reduce energy consumption is to reuse waste cold energy from industrial sources (e.g., liquefied natural gas) by an inertial carbon extraction system, a thermal swing process, or an external cooling loop. Baxter et al. [69,70] proposed a hybrid cryogenic carbon capture system via an external cooling loop (CCCECL), as illustrated in Fig. 10. The CCCECL process involves: 1) drying and cooling of flue gas from existing systems, 2) compressing and cooling of flue gas to a temperature slightly above the point where CO_2 forms a solid, 3) expanding the gas to further cool it, 4) precipitating an amount of CO_2 as a solid that depends on the final temperature, 5) pressurizing the CO_2, and 6) reheating the CO_2 and the residual flue gas via the incoming gases [69]. As a result, the CO_2 is captured in the liquid phase, and a N_2-rich stream is discharged. The CCCECL process has some configurations that can store energy in the form of LNG [70]. This capability manages the energy loss of CO_2 removal by using a stored refrigerant to drive the process during peak demand, transferring the reduced parasitic load to the grid to help meet demands, and regenerating the refrigerant during low-demand periods [69]. In addition, the rapid-load-change capability of CCCECL is beneficial in order to integrate conventional power generation systems with renewable intermittent power sources. The total energy consumption of the CCCECL route is less energy intensive (an average of 0.98 $MJ_{electrical}$/kg CO_2) than other conventional processes [71].

3.3. Anti-sublimation (AnSU)

An anti-sublimation CO_2 capture process (AnSU) was designed and proposed by Clodic and Younes [74]. As shown in Fig. 11, the whole process can be divided into five stages [19], including: 1) flue gas clean up, and cooling down to −40 °C with moisture removal; 2) heat exchange between rich flue gases and poor flue gases; 3) a refrigeration integrated cascade; 4) CO_2 freezing heat exchange; and 5) CO_2 recovery. The AnSU process has been demonstrated in a 660 MWe boiler with a CO_2 concentration of 15.47% at 60 °C and 120 kPa [72]. The energy penalty of the AnSU process is in the range of 3.8–7.2% of the power plant efficiency [73]. For comparison, retrofitting the MEA absorption process into a power plant would result in about a 14% decrease in the power plant efficiency, which is higher than that of AnSU [73].

Another advantage of the AnSU process is that defrosting the CO_2 (from the solid to liquid phase) on the heat exchanger surface allows recovery of the heat of fusion [72]. The latent heat of fusion is used to cool down the liquid blend of refrigerants before evaporation [74]. However, the moisture in the flue gas should be previously removed in order to prevent clogging and an unacceptably high rise in pressure during operation. In addition, the growing layer of frosted CO_2 on the surface of the heat exchanger would adversely affect the heat transfer, and thus, reduce the process efficiency. The materials used to make the heat exchangers have to be carefully selected with good thermal conductivity and mechanical stresses. It should also be noted that the freezing point of CO_2 is highly related to it partial pressure (i.e., concentration) in the flue gas. Although the frost point of pure CO_2 is −78.5 °C, it would decrease to −99.3 °C for flue gas (usually including 15% CO_2) from a typical coal-fired power plant [75]. Therefore, the total energy consumption in an AnSU process would increase along with the decrease in the initial CO_2 concentration in the flue gases, as well as the desired CO_2 capture efficiency [73].

3.4. Cryogenic distillation

Distillation is one of the most common separation technologies. A conventional cryogenic distillation process was proposed by Holmes and Ryan [76] for natural gas purification. The detailed separation process is summarized in Fig. 12. The feed gas is initially cooled by a pre-cooler, and then chilled to low temperature by the heat exchanger. The cooled feed gas is sent to the distillation column, which contains a number of vapor-liquid contact devices (such as trays or packing materials). The component steam after the distillation column is mainly divided into two parts: top and bottom products. The methane separated at the top is withdrawn via a partial condenser. Condensed CO_2 is gathered at the bottom of the distillation column. Part of the rich CO_2 stream is recycled to the distillation column by passing through the reboiler to provide vaporization heat. The other part of the CO_2 stream is further separated, and finally, the purified CO_2 product is extracted from the separator.

Despite its widespread industrial use and major benefits, the high energy demands of cryogenic distillation commonly contributes to over 50% of plant operating costs [77,78] Several energy-efficient distillation solutions based on process integration and intensification techniques have been proposed, such as cyclic distillation, a heat-integrated distillation column, reactive distillation, and thermally coupled columns [79,80]. Maqsood et al. [81–83] explored an intensified side-mounted and integrated switched cryogenic network configuration for CO_2 separation from natural gas. The investigation results showed a substantial reduction in the energy requirement; methane loss and size requirements were observed using the intensified hybrid cryogenic distillation networks [83]. The overall profit increased to 69.24% with the optimization of the intensified cryogenic network [84].

3.5. Controlled freezing zone

To mitigate the air pollution caused by coal combustion, some developing countries (e.g., China) have suggesting changing their energy resources from coal to natural gas [85]. However, the presence of acidic impurities (i.e., CO_2 and H_2S) in natural gas adversely affects its transport and combustion, which are necessary, so these compounds must be removed. Generally, the concentration of sour gas in raw natural gas is between 20% and 40%, with some concentrations as high

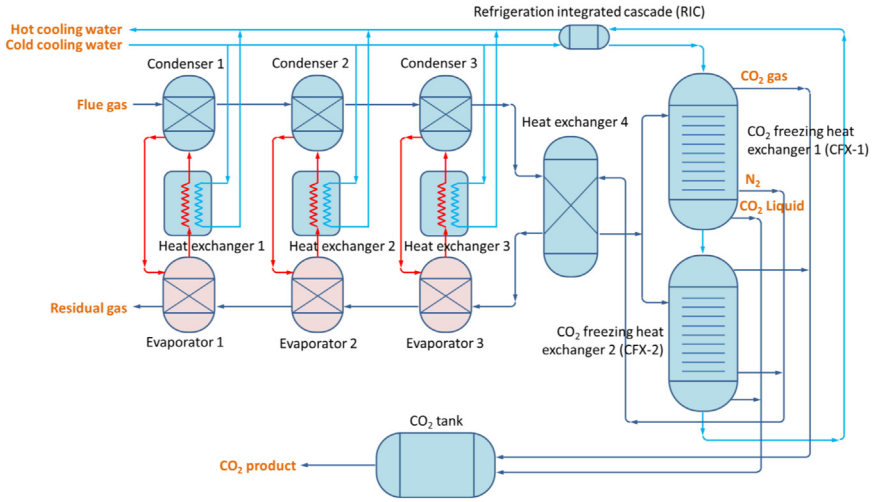

Fig. 11. Process flow diagram of the anti-sublimation CO_2 capture process (AnSU). [72].

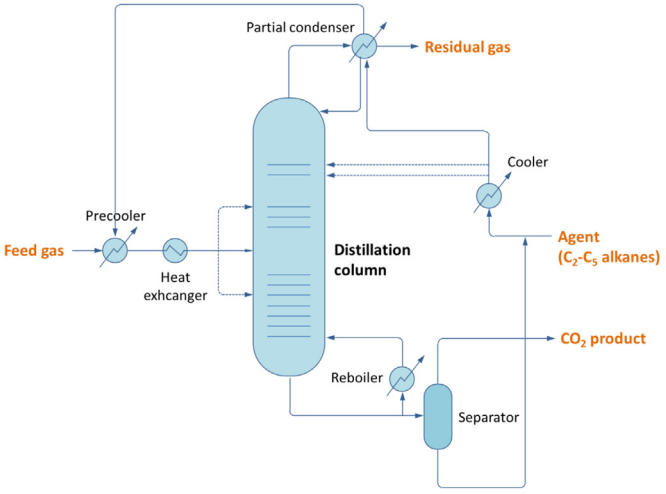

Fig. 12. The schematic flow diagram of Ryan Holmes process. [76].

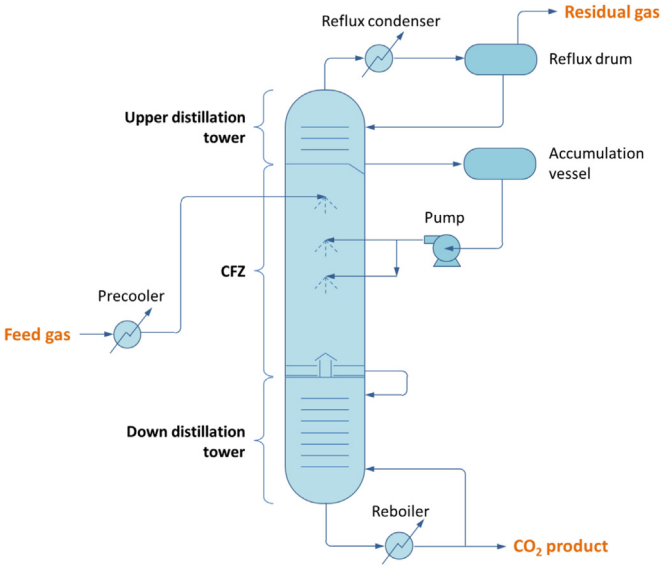

Fig. 13. Process flow diagram of the Controlled Freeze Zone (CFZ)™ process [88].

as 70%. The ratio of CO_2 to H_2S varies tremendously across geographic regions [86]. A controlled freezing zone (CFZ) process [87] provides an integrated solution to efficiently removal sour impurities from natural gas. As shown in Fig. 13, the whole process is preferably comprised of three sections, namely, an upper distillation section (UD), controlled freezing zone (CFZ), and down distillation (DD) section. During the separation process, the feed stream is first sent to a pre-cooler. Then, the chilled feed gas is introduced into the CFZ through a spray nozzle. The ExxonMobil Upstream Research Company has successfully demonstrated CFZ™ technology through a commercial demonstration plant in Wyoming [87]. Most of the captured CO_2 at ExxonMobil's commercial demonstration plant is used for enhanced oil recovery (EOR), and can add to the economic viability of producing sour natural gas reserves.

3.6. *CryoCell® process*

The CryoCell® process was developed by Cool Energy Ltd. and tested in collaboration with other industrial partners including Shell Global Solutions in Western Australia [89]. The configuration of the CryoCell® process is depicted in Fig. 14. The feed gas is initially dehydrated to low water specifications (5 ppm) so it can handle the downstream cryogenic operations. The dried gas then undergoes heat exchange with treated gas and cold CO_2 prior to cooling the CO_2 to its freezing point. The liquid is then expanded across a Joule-Thomson valve and enters the CryoCell® separator as a three-phase mixture. The solid CO_2 collected at the bottom of the separator is melted by a heater and separated as a liquid phase. The gas is compressed to sale-gas specifications, and the liquid is pumped to the required disposal pressure.

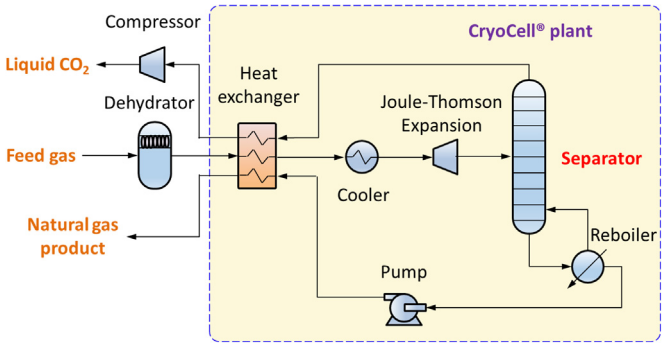

Fig. 14. Process flow diagram of the typical CryoCell® CO_2 capture process from natural gas [89].

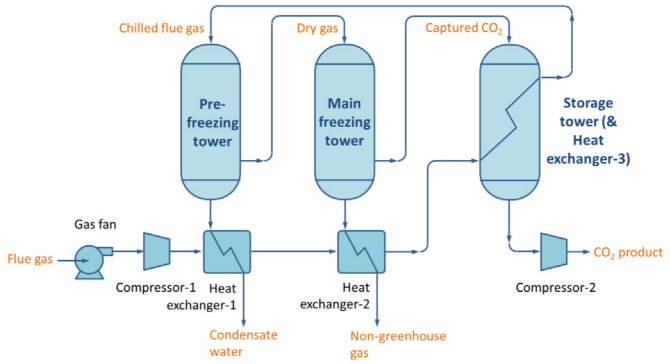

Fig. 15. The schematic of the SC based low temperature CO_2 capture process with heat recovery.

3.7. Stirling cooler system

Song et al. [94] designed a Stirling cooler system to capture CO_2 at low temperature with reasonable waste sensible and latent heat recoveries, as shown in Fig. 15. There are three parts (namely, a pre-freezing tower, main freezing tower and storage tower) of this system, and each part is chilled by a free-piston Stirling cooler. The pre-freezing tower is chilled by Stirling cooler-1 to separate H_2O in the flue gas by condensation. The additional heat in the main-freezing tower is removed by Stirling cooler-2 until the temperature is low enough to capture CO_2 by anti-sublimation. Furthermore, the frosted CO_2 gathers on the storage tower (chilled by Stirling cooler-3), and the residual gas is discharged to the ambient atmosphere. The detailed structure of this system can be found in reports by Song et al. [94,95,104]. The capture performance of the Stirling cooler-based cryogenic CO_2 capture system has been demonstrated on the laboratory scale by simulated flue gas (CO_2/N_2 binary gas). The total energy consumption can be controlled to values below 0.55 MJ $_{electrical}$/kg CO_2 [75]. However, so far, there has been no pilot or large-scale application of this method. For industrial CO_2 emissions (such as power plants, cement, steel and iron factories), the condition of the flue gas is far different from simulated gas (i.e., high flow rate, high temperature, dilute CO_2 concentration and more impurities) [90]. This is a critical challenge of the Stirling cooler-based cryogenic process and needs to be focused on in future work.

4. Advantages of cryogenic CO_2 capture technologies

4.1. High pressure and high purity CO_2 product

Following capture, utilization or storage are the typical paths for large amounts of CO_2. For both of these routes, compression and pipeline transport is necessary to deliver the produced CO_2 to designated sites. The purity of the CO_2 product is a key factor for transport, utilization and storage; especially for storage, as the site is usually far away from the CO_2 capture plant. High purity CO_2 can be easily compressed to the required pressure and then transported. During the cryogenic processes, CO_2 can be captured in different phases, e.g., liquid, solid or a combination (CO_2 slurry). Compared to the other separation technologies, CO_2 captured by cryogenic methods can achieve higher purity (over 99.9%) due to the inherent properties of desublimation. High purity CO_2 products can be more efficiently converted into valuable chemicals by catalytic or biological reactions (e.g., steam methane reforming and artificial photosynthesis), and can also be used in industrial food, fertilizer, etc. Another advantage of cryogenic processes is that an additional compression treatment can be avoided, and thus, the total energy penalty is reduced.

4.2. Energy storage

Another advantage of cryogenic CO_2 capture is that the low temperature CO_2 product could be reused as a potential cold energy source in industry (i.e., mechanical cleaning), and thus, it can be integrated with other low-temperature processes (i.e., natural gas liquefaction, etc.). Baxter's group [91,92] designed an energy and cost-efficient process, namely, energy storage for cryogenic carbon capture (CCC-ES), by combining a cryogenic process with an open natural gas (NG) refrigeration loop. The whole process can be divided into three stages: 1) Energy storing, where LNG production increases by 40% to use excess energy available on the grid. 2) Balanced, where LNG production equals the demand from the power plant to run the cryogenic carbon capture (CCC) process. 3) Energy recovery, where LNG production decreases by 70% to reduce the parasitic loss, and hence, place more power on the grid when demand is low.

5. Existing challenges and potential solutions

5.1. Cryogenic source

The advantages and limitations of the dominant low-temperature CO_2 capture technologies via different cold sources are summarized in Table 3. Because of the substantial energy requirements for cryogenic processes, the installation of cryogenic systems is highly limited by the available cryogenic sources. Although Liquefied Natural Gas (LNG) could be an ideal cold energy source, it also leads to a high dependency on the location, scale and other specifications of the natural gas factory. Compression and refrigeration are common methods used to provide cryogenic conditions. In addition to these, coolers are also one of the most dominant cryogenic sources. When the cooling in a cryogenic processes must be provided by refrigeration, the total energy requirement would increase. The coefficient of performance (COP) of a typical refrigerator that cools to −140 °C is typically 0.5, which means the consumed electricity would be equal to twice the thermal energy [66]. Heat recovery from the cold CO_2 products and residual gas could be a potential solution to reduce the energy consumption of compression and refrigeration. Thus, recuperative heat exchangers can be used to recover the sensible and latent heat from the residual streams and solid CO_2 during warming and melt processes [90,91].

5.2. Capture cost

The main challenge associated with the large-scale deployment of CCS is the capture cost. CO_2 capture plants and trains to carry compressed materials account for the majority of the whole CCS chain cost (i.e., 80% for capture and compression, 10% for transportation and 10% for storage) [96,97]. Raksajati et al. [98] and Siriwardhana et al. [99] indicated that, the CO_2 capture cost of a conventional MEA absorption process is approximately 55 USD$/ton CO_2. In contrast, the carbon tax in Australia and Europe are 23 AUD$/ton CO_2 and 3.5 €/ton CO_2, respectively, which seriously restricts the commercial application of CCS.

The capture performance of existing cryogenic processes is summarized in Table 4. The capture cost of a typical CO_2 capture process mainly consists of two parts: capital expenditure (CAPEX) and operational expenditure (OPEX). In cryogenic CO_2 processes, compressors and pumps, as well as multi-stream heat exchangers for CO_2 condensation and liquefaction, are the main energy-intensive units [100,101]. Compared to traditional absorption or adsorption process, the CAPEX of cryogenic methods is low due to their smaller equipment sizes (without use of a large absorption/adsorption tower). Meanwhile, the OPEX of cryogenic processes also decreases because solvent/sorbent regeneration costs are avoided. However, it should be noted that different types of energy types are required for different CO_2 capture technologies [94,102]. For the mature amine absorption techniques,

Table 3
Advantages and limitations of the existing low temperature CO_2 capture technologies.

Category	Cold energy source	Advantages	Limitations	Refs.
Packed bed	Liquid nitrogen gas (LNG)	◆ Atmosphere ◆ Simultaneous H_2O and CO_2 removal ◆ Avoiding high pressure drop ◆ surface area-to-volume ratio of the column	◆ Depends on the availability of LNG ◆ Lab scale	[66,93]
AnSU	Liquefied natural gas (LNG)	◆ Atmosphere ◆ Lower energy penalty than MEA absorption ◆ Pilot demonstration	◆ Depends on the location of natural gas station ◆ No H_2O can be tolerated ◆ Frost CO_2 adversely affects heat conduction ◆ undesired mechanical stresses	[72]
CryoCell	Chiller	◆ No process heating system required ◆ No corrosion potential ◆ No foaming potential ◆ Avoid compression cost	◆ More suitable for high CO_2 concentration (higher than 20%) ◆ High compression power requirement	[89]
Distillation	Compressor and cooler	◆ Avoid compression cost ◆ Easy to be pumped to storage site ◆ Energy storage potential ◆ Water saving potential ◆ Simultaneous removal of other pollutants (Hg, SO_x, NO_2, HCl, etc.)	◆ Capital cost for pressure difference ◆ High installation cost	[69]
Stirling cooler	Striling cooler	◆ Atmosphere ◆ Simultaneous H_2O and CO_2 removal ◆ Lower energy penalty than MEA absorption ◆ Energy storage potential	◆ Exergy loss due to temperature difference ◆ Difficulty of frost layer scrapping ◆ Lab scale	[94,95]

two aspects of the energy should be considered. One part is the thermal energy used for solvent regeneration, which is the dominant energy penalty of the whole process. The other small part is the electrical energy used to operate the installations. For the cryogenic processes, the cryogenic conditions and operation of the system are both based on electrical energy consumption when low-cost cold energy (e.g., liquefied natural gas) is unavailable. The conversion ratio between thermal and electoral energy is approximately 0.4 [75].

5.3. Efficiency

The CO_2 capture efficiency of cryogenic processes highly depends on the operating temperature. With decreasing temperature, the efficiency obviously increases. At cryogenic temperatures, the exhaust exiting the capture units could contain less CO_2 than the ambient air [105]. The heat exchanger networks (HENs) also have a critical influence on the efficiency of cryogenic CO_2 capture processes. Well-designed HENs (with optimal viscosity and thermal conductivity) can significantly facilitate a decrease in the destruction of exergy [100,106].

The temperature difference between the cryogenic system and the ambient conditions would also lead to heat loss, and thus, decrease the total efficiency. A vacuum interlayer would be efficient an approach to reduce exergy loss [104]. The material of the heat exchangers should be carefully selected to operate at dramatically temperatures, while maintaining good heat conductivity and mechanical stresses. The frosted CO_2 on the surface of the heat exchanger may adversely affect heat transfer between the gas and solid phase [73]. For the CO_2 desublimation process, the frosted CO_2 on the heat exchanger plays an important role as the heat transfer medium and affects subsequent CO_2 deposition. Although many reports have investigated frost formation processes, most of them are related to moisture [107]. The frost behavior of CO_2 is different from moisture, and the whole process can be divided into three periods [108]: (1) Crystal growth, (2) Frost growth, and (3) Frost formation. It is also worth noting that the presence of other components (e.g., N_2, O_2 and H_2O) would also affect the CO_2 frosting process. Therefore, CO_2 frost from flue gas involves an intricate deposition process compared to that of pure water or CO_2, and further efforts would be necessary.

5.4. Impurities

The flue gas often contains several impurities, such as SO_x, NO_x, particulate matter (PM) and mercury, etc. The presence of trace impurities in the produced CO_2 has a negative impact on its pipeline transportation, geological storage and/or Enhanced Oil Recovery (EOR) applications. All of these negative influences require stringent quality guidelines for the product's application, as well as purification of the CO_2 stream prior to exposing it to any of these applications [109].

It is well known that water forms ice under cryogenic conditions,

Table 4
Comparison of the capture performance for the existing low temperature CO_2 capture technologies.

Cryogenic processes	Gas sources	CO_2 recovery	Energy consumption	Phase state	Refs.
Cryogenic packed bed	Coal-fired power plants (10 vol% CO_2, 89 vol% N_2, 1 vol% H_2O)	99.%	1.8 $MJ_{electrical}$/kg CO_2	Liquid	[66]
AnSU process	Coal-fired power plant (12% CO_2)	90%	1.18 $GJ_{electrical}$/ton CO_2	Liquid	[19]
CryoCell process	Raw natural gas (20–35 mol% CO_2)	–	–	Liquid	[89]
CFZ	Raw natural gas (8–71% CO_2)	98–99% CH_4	–	Liquid	[103]
CCC-ECL	Raw natural gas (32.5 mol% CO_2)	95.6 mol%	1.401 kWh/kg CO_2	Liquid	[78]
Stirling Cooler system	Coal-fired power plants (13% CO_2 and 87% N_2)	85%	3.4 $MJ_{thermal}$/kg CO_2	Solid	[104]

which can lead to the clogging of pipes and equipment. To achieve a high capture efficiency in cryogenic processes, any moisture should be removed by the pretreatment units. The cryogenic packed bed process designed by Tuinier et al. [17] could overcome the related bottleneck based on differences in the dew and sublimation points of different compounds. H_2O and CO_2 can be condensed and desublimated, respectively, at different positions on the packed bed.

In addition, acidic impurities can lead to corrosion of the heat exchangers and the entire installation. Normally, an efficient desulfurization and DeNOx treatment should be conducted prior to cryogenic CO_2 separation, which would lead to an increase in the capture cost. To mitigate the negative influence of the impurities, some researchers have tried to design novel cryogenic CO_2 capture processes, which could simultaneously remove other pollutants, such as SO_x, NO_x, PM and mercury. The cryogenic process proposed by Fazlollahi et al. [110] provided an innovative route to desublimate CO_2 and condense other pollutants, removing all species that are less volatile than CO_2.

6. Prospective

6.1. Hybrid with other technologies

To overcome bottlenecks in the conventional technologies, the combination of two or more standalone CO_2 capture methods to create hybrid processes (e.g., absorption-membrane, adsorption-membrane, cryogenic-adsorption, and cryogenic-membrane, etc.), has attracted increasing attention [14]. The potential arrangement of hybrid CO_2 capture processes can be classified into four categories, absorption-based, adsorption-based, membrane-based and cryogenic-based processes [111]. Due to their advantages in CO_2 recovery and purity, cryogenic technologies are desirable candidates to create hybrid processes with other techniques (i.e., cryogenic-absorption, cryogenic-adsorption, cryogenic-membrane and cryogenic-hydrate, etc.). Merkel et al. [127] and Scholes et al. [112] proposed and optimized a novel hybrid process that incorporates three membrane stages with low temperature separation, as illustrated in Fig. 16. The designed hybrid process without Flue Gas Desulfurization (FGD) and de-NOx could achieve a parasitic load of 31–34% and a cost of capture of US$ 31–32 per ton of CO_2 [112]. If FGD and de-NOx are required, the capture cost would increase to US$ 41–42 per ton of CO_2 [112]. This capture cost is competitive with state-of-the-art MEA solvent technology for CO_2 capture from a brown coal power station [112]. Belaissaoui et al. [113] proposed a novel CO_2 capture process combining membrane and cryogenic units, as shown in Fig. 17. The simulation results demonstrated that for a CO_2 feed concentration between 15% and 30%, the total energy requirement was lower than 3 $MJ_{thermal}$/kg CO_2 (including the compression of CO_2 to 110 bar), with a CO_2 recovery ratio above 85% and CO_2 purity above 89%. This value showed a reduced energy requirement compared to the reference MEA absorption technology

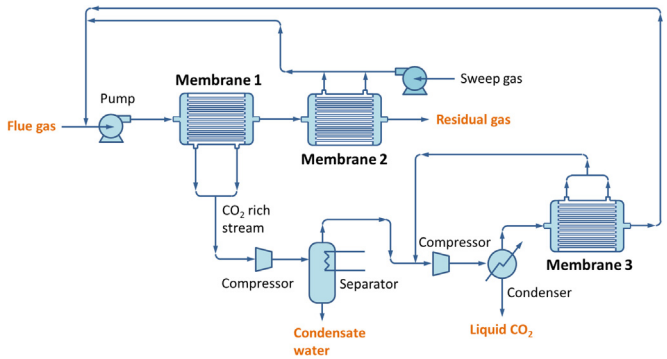

Fig. 16. The membrane-low temperature post combustion CO_2 capture process. [112].

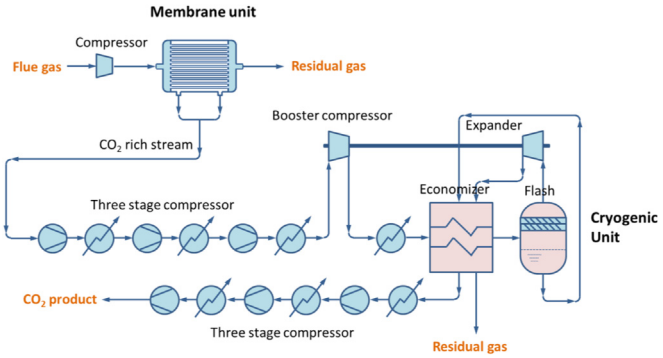

Fig. 17. The membrane-cryogenic hybrid CO_2 capture process [30].

[113].

The performances of existing cryogenic-based hybrid processes are summarized and compared in Table 5. Cryogenic-based hybrid processes are one of the most promising alternatives, and could capture CO_2 as a compressed liquid product from coal-fired power plant with a low energy consumption to 1.163 MJ/kg CO_2 [120]. According to results from Belaissaoui et al. [30], with a CO_2 concentration in the range of 11–30%, the energy consumption by a cryogenic-membrane hybrid process varied from 2.0 to 4.0 MJ/kg CO_2, which is lower than that of MEA absorption (4.0 MJ/kg CO_2), single-stage membrane separation (2.0–6.0 MJ/kg CO_2) and a standalone cryogenic process (2.5–12 MJ/kg CO_2).

6.2. Combined with CO_2 utilization

To safely and permanently store the captured CO_2, the availability and capacity of storage sites (e.g., deep saline aquifers/DSA, depleted oil and gas fields/DOGF, etc.) are key factors that need to be comprehensively evaluated. Although the capacity (globally approximated at 8000 Gton and 55,000 Gton) of the potential storage sites is sufficient compared to the huge amount of CO_2 emission (32.4 Gton globally in 2014, and approximately 14 Gton from coal combustion), costs vary significantly from €1–7/ton CO_2 stored for onshore DOGF to €6–20/ton for offshore DSA. [121–123]. Therefore, in addition to storage, CO_2 utilization has received more and more attention in recent years [124,125]. CO_2-EOR is a proven technology for increasing oil recovery and simultaneously storing CO_2 permanently in the subsurface of the earth. Commercial CO_2 capture and utilization (CCU), such as mineralization and CO_2-EOR, can make a positive contribution to the reduction of climate change by providing long-term CO_2 storage and an economic drive for CO_2 capture and network development. Other CCU propositions (e.g., converting CO_2 into value-added chemicals via catalysis or bio-synthesis, etc.), although involving smaller quantities of CO_2 without long-term storage potential, can also provide economic drivers for CO_2 carbon capture, especially in locations where access to storage sites is limited.

Nevertheless, the composition of flue gas is complex (including particulate matter, SOx, and NOx, etc.) and the CO_2 concentration in the gas is usually dilute; thus, pretreatment and concentration are necessary to ensure efficient utilization. By combining cryogenic capture with utilization technologies, high purity CO_2 (liquid or solid) can be easily obtained and used for cereal preservation (bactericide), additives to beverages, food packaging/conversion, dry-cleaning, extraction, mechanical industries, fire extinguishers, air-conditioning and water treatment [126]. The high quality CO_2 collected via cryogenic separation could also be an ideal feedstock for microorganisms (e.g., microalgae) to produce high value-added compounds (polysaccharides, pigments, amino acids, vitamins and polyunsaturated fatty acids, etc.), which could be used for health care, functional foods, antioxidant additives and so on. This multitude of potential applications is another

Table 5
Summary of existing cryogenic based hybrid CO_2 capture processes.

Hybrid processes	Gas sources	Product purity	Energy consumption	Phase state	Refs.
Cryogenic-membrane	Coal-fired power plants	98.3 mol%	1.215 GJ/t CO_2	Liquid	[114]
	Biogas	99.9% CH_4	–	Liquid	[115]
	Coal-fired power plants	65%–67%	48 $€_{2008}$/t CO_2	Liquid	[116]
	Coal-fired power plants	94.1%	1.249 GJ/t CO_2	Liquid	[117]
Cryogenic-hydrate	IGCC power plant	95 mol%–97 mol% CO_2	–	Liquid	[118]
Cryogenic-adsorption	Raw natural gas	99.9992% CO_2 and 99.9995% CH_4	–	Gas	[119]
Cryogenic-absorption	Coal-fired power plants	93.8 wt%	1.163 GJ/t CO_2	Gas	[120]

unique, superior property for cryogenic-based CO_2 capture processes.

7. Conclusion

The development of low-temperature CO_2 capture from large industrial CO_2 emissions is summarized in this review. One of the main advantages of cryogenic or cryogenic-based CO_2 capture processes is that high purity CO_2 product can be obtained in either a compressed liquid or solid phase, which is beneficial for transport, utilization and storage. With respect to environmental protection, cryogenic technologies are also a green method, as CO_2 could be separated from industrial stations without organic solvents and sorbents using. However, it should be noted that cryogenic technologies are only more competitive when low-cost cold energy sources (e.g., liquefied natural gas) are available. Furthermore, the destruction of exergy due to dramatic temperature variations is also a critical challenge for the commercial application of cryogenic processes. Therefore, more efficient cryogenic processes should be designed and optimized in the future.

Acknowledgements

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